



# The influence of particle size on the potential of enhanced basalt weathering for carbon dioxide removal - Insights from a regional assessment

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## ARTICLE INFO

Handling editor: Zhifu Mi

### Keywords:

Negative emissions  
Basalt weathering rates  
Carbon dioxide removal  
Enhanced weathering  
Soil amendment  
Shrinking core model

## ABSTRACT

Enhanced weathering through basalt application on agricultural land represents a proposed strategy for the removal of carbon dioxide from the atmosphere. It has been shown that enhanced weathering is principally feasible on a global scale, but there is still uncertainty with respect to the predicted drawdown in a given timeframe. This information is however vital to evaluate, if enhanced weathering should be further considered as a factor to alleviate the impact of the climate crisis. With this in mind, this article reviews of the current state of research and estimates the CO<sub>2</sub> drawdown for scenarios using basalt powders of different particle size distribution (<100 µm, <10 µm and <1 µm). Calculated with a modified shrinking core model, the amount of powder dissolved within a timeframe of 10 years is approximately 16% (<100 µm), 55% (<10 µm) and 99.9% (<1 µm). This corresponds to a gross CO<sub>2</sub> removal of 0.045 t CO<sub>2</sub> t<sup>-1</sup> of rock (<100 µm) and 0.153 t CO<sub>2</sub> t<sup>-1</sup> of rock, (<10 µm). We evaluate our results on regional scale through a case study for Austria, including emissions from mining, comminution, application and transport. Assuming an average distance of 300 km from mine to field, the net CO<sub>2</sub> drawdown decreases to approximately 0.027 t CO<sub>2</sub> t<sup>-1</sup> of rock (<100 µm) or 0.096 t CO<sub>2</sub> t<sup>-1</sup> (<10 µm), when rail transport is used. For truck transport, the numbers are reduced to -0.030 t CO<sub>2</sub> t<sup>-1</sup> of rock (<100 µm) or 0.039 t CO<sub>2</sub> t<sup>-1</sup> (<10 µm), respectively. Accordingly, at the current CO<sub>2</sub> intensity, transport related emissions may cancel out any drawdown if grain sizes (<100 µm) are used. Our estimates suggest that enhanced weathering will only significantly contribute to net CO<sub>2</sub> drawdown if grain sizes (<10 µm) are used. Under these conditions the large-scale application of particles with a diameter <10 µm may remove about 2% of Austria's annual Greenhouse gas emissions. We discuss challenges towards this goal, including the enormous amounts of rock needed and the energy requirement related to grinding, as well as uncertainties related to actual field weathering rates. Those uncertainties hinder the precise quantification of CO<sub>2</sub> drawdown as of now. While enhanced weathering remains a promising path for climate change mitigation, further research at laboratory and field scale is required to put this technology to optimal use.

## 1. Introduction

Within the framework of the Paris Agreement, the signing nations have agreed to keep global warming below 2 °C with respect to the pre-industrial level and to pursue efforts to limit the increase to 1.5 °C above pre-industrial levels to mitigate the effects of human caused climate change. Estimates of the remaining global carbon budget consistent with a 1.5 °C and 2 °C scenario imply that the annual global output of total emissions should be curbed down to 25 GtCO<sub>2</sub>e (carbon dioxide equivalent) and 41 GtCO<sub>2</sub>e by the year 2030, respectively (Rogelj et al., 2018). However, within the framework of current policies, the estimated

annual output in 2030 will be around 60 GtCO<sub>2</sub>e and even if countries manage to reach their self-determined contributions to reduce emissions, this value will only lower to about 54 GtCO<sub>2</sub>e (Emissions Gap Report, 2019). With the remaining carbon budget rapidly diminishing, negative emission technologies (NET) are increasingly coming into focus. Instead of reducing CO<sub>2</sub> production, NETs focus on removing CO<sub>2</sub> from the atmosphere and storing it in oceanic and terrestrial sinks. Current approaches include bioenergy with carbon capture and storage (BECCS), direct air capture (DAC), Enhanced Rock Weathering (EW), afforestation, ocean fertilization and the conversion of biomass into biochar for soil amendment (Negative Emissions Technologies and

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<https://doi.org/10.1016/j.jclepro.2021.128178>

Received 8 January 2021; Received in revised form 28 May 2021; Accepted 29 June 2021

Available online 2 July 2021

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Reliable Sequestration: A Research Agenda, 2019; Smith et al., 2016). Large scale deployment of negative emission technologies within the 21st century is an integral part of the vast majority of models, consistent with the 1.5 °C and 2 °C scenario (Workman et al., 2020). These models often imply large scale removal of atmospheric CO<sub>2</sub> by NETs only after 2050, allowing a temporal overshoot of our remaining carbon budget (Hilaire et al., 2019). It has been argued that within the framework of the current policies it might be impossible to reach the 1.5 °C goal without relying on negative emission technologies (Anderson et al., 2020; Hickel and Kallis, 2020). Until now different NETs are at different stages of development (Anderson and Peters, 2016), and at the moment there is no single solution for the removal of CO<sub>2</sub> at scale ready for deployment (Fuss et al., 2018). This has led to a more general debate whether NETs provide promising avenues for achieving the goals of the Paris Agreement (Schlesinger and Amundson, 2019). Provided the uncertainties on a global scale and with the only alternative being a more rigorous cut in emissions, it is required to precisely quantify the future potential of NETs and how they can be incorporated into climate change mitigation scenarios (Anderson and Peters, 2016; Fuss et al., 2014).

CO<sub>2</sub> consumption through reaction with mafic and ultramafic rocks and minerals is one of the technologies which has already been proposed 30 years ago as an industrial process (Lackner et al., 1995; Seifritz, 1990; see Kelemen et al. (2020) for an up to date review of the topic), and the potential through diffuse spreading of rock powder to the land surface has been reviewed by Schuiling and Krijgsman, (2006). This technology is referred to as Enhanced Rock Weathering (ERW), enhanced weathering (EW), or Terrestrial Enhanced Weathering (TEW). We use the term EW in this study. The concept has since been investigated in a number of different scenarios. The potential of mine tailings from mafic and ultramafic deposits has been evaluated (e.g. Assima et al., 2014, 2012; Gras et al., 2020; Hamilton et al., 2018; Harrison et al., 2013; Lechat et al., 2016; Oskierski et al., 2016; Power et al., 2020; Pronost et al., 2011; Thom et al., 2013; Wilson et al., 2014). Investigations include numerical modeling studies on global (Hartmann et al., 2013; Köhler et al., 2010; Moosdorf et al., 2014; Streffler et al., 2018; Taylor et al., 2016) and regional scale (Aviso et al., 2021; Lefebvre et al., 2019; Renforth, 2012). While some studies focus on the application to croplands (Beerling, 2017; Haque et al., 2019; Kantola et al., 2017; Taylor et al., 2017) others focus on marine environments and the effect of EW on ocean alkalinity (Bach et al., 2019; Hangx and Spiers, 2009; Montserrat et al., 2017; Renforth and Henderson, 2017; Rigopoulos et al., 2018a). Additionally, several field and lab studies have been carried out so far (Amann et al., 2020; Dietzen et al., 2018; Haque et al., 2020a, 2020b; Kelland et al., 2020; Renforth et al., 2015; ten Berge et al., 2012).

One advantage of EW is relatively low land use compared to other NETs and low use of the resource water (Smith et al., 2016). Moreover, enhanced weathering of dunite and basalt is cost competitive with respect to other methods, ranging from 60 US \$ per t CO<sub>2</sub> for dunite to 200 US \$ for basalt (Streffler et al., 2018). While the relatively high

energy input required for EW related to mining, milling and transport of the rocks reduces efficiency, the CO<sub>2</sub> drawdown of olivine may still reach a net value of 0.5–1 t CO<sub>2</sub> per t of rock (Beerling et al., 2018; Moosdorf et al., 2014). Olivine is used in many studies related to EW for its high CO<sub>2</sub> drawdown relative to rock mass (e.g. Dietzen et al., 2018; Renforth et al., 2015, Table 1), but the potential release of toxic metals during dissolution is a disadvantage (Amann et al., 2020). In contrast, Haque and coworkers showed in a number of studies that wollastonite (CaSiO<sub>3</sub>) amendment of soil is beneficial for crop yield, while at the same time increasing soil inorganic carbon content (Haque et al., 2019, 2020a, 2020b). Similarly, volcanic rock is already used for soil amelioration and results from an experimental study on the use of basalt powder, added to slightly acidic clay-loam soil, show cumulative removal of 4 t CO<sub>2</sub> ha<sup>-1</sup> within 5 years (Kelland et al., 2020). This study also found an increasing crop yield (sorghum) by 21 ± 9.4%. In a recent study Beerling et al. (2020) indicated the potential of basalt application to cropland for CO<sub>2</sub> removal on a global scale. They estimated the potential of CO<sub>2</sub> removal for application of ground basaltic rock on cropland in the range of about 0.5–2 GtCO<sub>2</sub> y<sup>-1</sup>. The aggregate amount of carbon dioxide removal, if sustained over 50 years could be between 25 and 100 Gt CO<sub>2</sub>. The study also showed the potential for removal of CO<sub>2</sub> in temperate climate regions. Factors influencing the potential of EW include climate, agricultural area and practice, availability of suitable rocks, available transport networks, the CO<sub>2</sub> intensity of electricity generation, and CO<sub>2</sub> emissions from mining, comminution and transport. The importance of transport distance has been addressed by Lefebvre et al. (2019). The authors assessed the potential of basalt for EW in a part of Brazil and concluded that the maximum transport between the quarry and field would 990 ± 116 km, above which the transport related CO<sub>2</sub> emissions would offset the potential CO<sub>2</sub> capture. Therefore, if EW is to be used in the near future, the regional availability of suitable resources is a prerequisite and regionally tailored approaches are needed to put the technology to optimal use.

Here we selected Austria as suitable case example to evaluate the feasibility of EW in a regional context. Towards the realization of the European Green Deal, countries with an efficient railway network, a wide variety of lithologies within reach, and relatively high share of hydropower for electricity generation, like Austria, at first sight seem to be well suited for EW as a technology. However, before EW can be applied, it remains to be quantified whether enough suitable rocks are available in Austria or bordering countries and whether the energy demand for mining, transport and mineral comminution and consequent CO<sub>2</sub> production substantiate enhanced basalt weathering as a reliable strategy. In the following, we will first review the current state of research in the field before we will use our case study to demonstrate how the feasibility of EW can be evaluated in a regional setting.

**Table 1**

Theoretical CO<sub>2</sub> drawdown (R<sub>CO<sub>2</sub> max</sub> and R<sub>CO<sub>2</sub> low</sub>) in kg CO<sub>2</sub> t<sup>-1</sup> of rock (mineral) for common basalt forming minerals and basalt. R<sub>CO<sub>2</sub> max</sub> is based on congruent dissolution and R<sub>CO<sub>2</sub> low</sub> is based on incongruent dissolution. Bold numbers indicate R<sub>CO<sub>2</sub> low</sub> and R<sub>CO<sub>2</sub> max</sub> used during modeling, based on the basalt composition given in Navarre-Sitchler and Brantley (2007).

Name	Formula	g mol <sup>-1</sup>	R <sub>CO<sub>2</sub> max</sub>	R <sub>CO<sub>2</sub> low</sub>
Forsterite	Mg <sub>2</sub> SiO <sub>4</sub>	140.7	1251	1251
Enstatite	Mg <sub>2</sub> Si <sub>2</sub> O <sub>6</sub>	200.8	877	877
Diopside	CaMgSi <sub>2</sub> O <sub>6</sub>	216.5	813	813
Hornblende	(Ca <sub>0.6</sub> Na <sub>0.3</sub> K <sub>0.1</sub> ) <sub>2.5</sub> (Mg <sub>0.6</sub> Fe <sub>0.3</sub> Al <sub>0.1</sub> ) <sub>5</sub> [(OH) <sub>0.75</sub> F <sub>0.25</sub> ] <sub>2</sub> (Si <sub>0.75</sub> Al <sub>0.25</sub> ) <sub>2</sub> Si <sub>6</sub> O <sub>22</sub>	868.4	811	507
Orthoclase	KAlSi <sub>3</sub> O <sub>8</sub>	278.3	632	158
Anorthite	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	278.2	1265	316
Leucite	KAlSi <sub>2</sub> O <sub>6</sub>	218.3	807	202
Nepheline	Na <sub>3</sub> KAl <sub>4</sub> Si <sub>4</sub> O <sub>16</sub>	584.3	1205	301
Basalt	Si <sub>1</sub> Ti <sub>0.025</sub> Al <sub>0.329</sub> Mg <sub>0.310</sub> Fe(III) <sub>0.02</sub> Fe(II) <sub>0.193</sub> Ca <sub>0.273</sub> Na <sub>0.061</sub> K <sub>0.007</sub> O <sub>3.394</sub>	123.5	968	429
Basalt	Ca <sub>0.3</sub> Mg <sub>0.1</sub> Fe <sub>0.4</sub> Al <sub>0.3</sub> SiO <sub>3.25</sub>	125.0	<b>880</b>	<b>282</b>

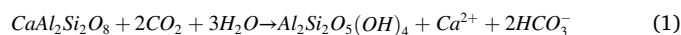
## 2. Theoretical background and methodology

### 2.1. Rock powder for soil amelioration

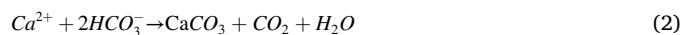
One of the goals proclaimed in the Paris agreement on climate change is to “[...] foster climate resilience and low greenhouse gas emissions development, in a manner that does not threaten food production”. In this context, the potential co-benefits of soil amelioration through application of basaltic rocks to croplands are noteworthy. They largely contrast the societal friction points, related to competition for land use, connected to BECCS, afforestation and reforestation. In addition, EW could be co-deployed for the two latter technologies (Amann and Hartmann, 2019; Kantola et al., 2017). Application of silicate–rock fertilizers to agricultural land is not a new concept and it has been investigated already by e.g. Hensel (1894) or more recently by e.g. Van Straaten (2006). The potential benefits are linked to the fact that silicon is actively accumulated in a number of crops including wheat, barley and sugar beet (Guntzer et al., 2012). Benefits of silicon accumulation are especially important under environmental stress because silicon alleviates toxicity of some toxic metals, improves Potassium, Phosphor and calcium uptake, alleviates the effects of drought and increases resistance to pathogens and insects (Adrees et al., 2015; Guntzer et al., 2012). The recycling of plant silicon in unaltered ecosystems accounts for a high percentage in the silicon pool, owed to the re-dissolution of plant phytoliths. Under agricultural use, however, this cycle is interrupted due to the removal of crops. Especially with silicon accumulating plants this can warrant silicon fertilization (Savant et al., 1997), and a positive effect on yield has been reported for instance in the case of wheat with a grain yield increase of 4.1–9.3% (Liang et al., 1994). Soil amendment through application of basalt has positive effects on the cation exchange capacity in soils and supplies potassium, phosphor and calcium (Gillman et al., 2002). In addition aluminum toxicity is a problem with acid soil conditions around the world. Silicon in general and basalt in particular have been shown to effectively decrease aluminum toxicity in acid soils (Cocker et al., 1998; Shamshuddin and Kapok, 2010). For these potential co-benefits, basaltic rock may be the best candidate for EW through application on agricultural land, despite its relatively low capability to sequester CO<sub>2</sub>, compared to dunite. Moreover release of Chromium, Nickel and Cobalt from dunite (Renforth et al., 2015; ten Berge et al., 2012) and serpentinites (Kierczak et al., 2016) and potential accumulation in soil and crops may counteract the intended soil amelioration and the same is true for low Ca/Mg ratios (Echevarria, 2018). While serpentinites and amphiboles might exhibit good reactivity, asbestos group minerals are generally a safety concern during mining, milling, and spreading of rock powder, thereby even if handled in a safe way, public perception could impose barriers on their application (Gwenzi, 2020).

### 2.2. CO<sub>2</sub> drawdown through weathering

Over geological time scales, atmospheric carbon dioxide is affected by the weathering of silicate rocks through the transformation of CO<sub>2</sub> into HCO<sub>3</sub><sup>−</sup> (Amiotte Suchet et al., 2003; Berner et al., 1983; Dessert et al., 2003; Gaillardet et al., 1999; Gislason et al., 2009; Walker et al., 1981). For instance, the rise of the Himalayas and consequent increased weathering has resulted in CO<sub>2</sub> drawdown and a general cooling of global climate (Raymo and Ruddiman, 1992). Similarly, past cooling phases of the climate since the Archean could be directly linked to the drawdown of CO<sub>2</sub> from the atmosphere (e.g. Elliot Smith et al., 2008; Kump et al., 1999; Lowe and Tice, 2004). The idea behind EW is to speed up this process through grinding of rocks and targeted application to increase weathering rates. Exemplarily the chemical reaction can be followed through incongruent dissolution of anorthite (eq. (1)). The dissolution of primary silicates leads to formation of secondary precipitates, releasing cations and transforming CO<sub>2</sub> into HCO<sub>3</sub><sup>−</sup>.



Ideally the HCO<sub>3</sub><sup>−</sup> ion will subsequently be transported to the ocean (Renforth and Henderson, 2017), where it possibly partially alleviates ocean acidification (Hartmann et al., 2013; Taylor et al., 2016). If supersaturation with respect to individual carbonate phases is reached, solid carbonates might form (eq. (2)).



Carbonate formation is an important mechanism for the in situ fixation of CO<sub>2</sub> through Carbon Capture and Storage (For a review of mineral carbonation in general see e.g. Kelemen et al., 2020; Snæbjörnsdóttir et al., 2020). However, the aim of enhanced weathering is to convert CO<sub>2</sub> into alkalinity, as the formation of carbonates will reduce the processes efficiency (eq. (2)).

The maximum amount of CO<sub>2</sub> drawn from the atmosphere through silicate dissolution is a function of the cation flux (mostly Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup> and Na<sup>+</sup>) which is charge balanced by the formation of HCO<sub>3</sub><sup>−</sup>. Drawdown potential can be expressed as the amount of CO<sub>2</sub> (R<sub>CO2</sub>) removed from the atmosphere per mass of rock (t CO<sub>2</sub> t<sup>−1</sup> of rock). This value might be reduced during riverine transport (Hotchkiss et al., 2015; Marx et al., 2017; Polsenaere et al., 2013) and it will further decrease, depending on the apparent carbonate equilibrium in the ocean (Beerling et al., 2020). For the relatively fast weathering of Ca and Mg bearing silicates, R<sub>CO2</sub> is sometimes based solely on Mg<sup>2+</sup> and Ca<sup>2+</sup> contents (Renforth, 2012). In the case of basalt the contribution of monovalent cations (Na<sup>+</sup> and K<sup>+</sup>) needs to be included, as they are frequently present in mafic and ultramafic volcanic rocks (see Table 1) in the form of relatively fast weathering minerals such as nepheline (Tole et al., 1986). In addition, the calculation of R<sub>CO2</sub> from incongruent dissolution, is based on the idea of aluminum conservation through the formation of secondary minerals (eq. (1)). However, congruent dissolution of the aluminosilicate will precede the formation of the secondary phase (Maher et al., 2009; Steefel and Van Cappellen, 1990) (eq. (3)), and far from equilibrium conditions can be sustained during basalt weathering for instance through complexation of Al<sup>3+</sup> with organic acids (Perez-Fodich and Derry, 2019).

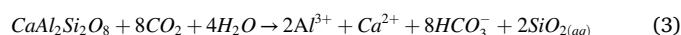


Table 1 provides the R<sub>CO2</sub> for minerals commonly found in basalt and two different compositions of basalt (Gudbrandsson et al., 2011; Navarre-Sitchler and Brantley, 2007). R<sub>CO2 low</sub> is based on incongruent dissolution, where CO<sub>2</sub> is charge balanced against the cations released during dissolution (ΣCO<sub>2</sub> = meq(Ca<sup>2+</sup> + Mg<sup>2+</sup> + Na<sup>+</sup> + K<sup>+</sup>)). R<sub>CO2 max</sub> is based on congruent dissolution including Al<sup>3+</sup>, Fe<sup>3+</sup>, Fe<sup>2+</sup> and Ti<sup>4+</sup> in the calculation. The latter process has also been proposed in (Navarre-Sitchler and Brantley, 2007) to assess CO<sub>2</sub> drawdown from basalt weathering. The values of 282 kg t<sup>−1</sup> of basalt (R<sub>CO2 low</sub>) and 880 kg t<sup>−1</sup> of basalt (R<sub>CO2 max</sub>) will be used for evaluation of the potential in a pessimistic and optimistic scenario in this study.

### 2.3. Basalt weathering rates in the context of enhanced weathering

The dissolution of basaltic glass at far from equilibrium conditions has been investigated in mixed flow reactors by (Galeczka et al., 2014; Gislason and Oelkers, 2003; Oelkers and Gislason, 2001; Stockmann et al., 2011; Wolff-Boenisch et al., 2004). These studies show a minimum dissolution rate in the circumneutral pH area (see Fig. 1), commonly observed for aluminosilicates (Gautier et al., 1994; Gudbrandsson et al., 2014). In contrast, Al-free silicate minerals, such as enstatite or forsterite exhibit a near linear dependence of dissolution rates on pH (Fig. 1). Accordingly, the respective mineralogical composition of a basaltic rock may influence the stoichiometry of the released cations. This has been shown in experiments, where a U-shaped release of Ca<sup>2+</sup> as a function of pH was shown, connected to the dissolution of plagioclase, while for

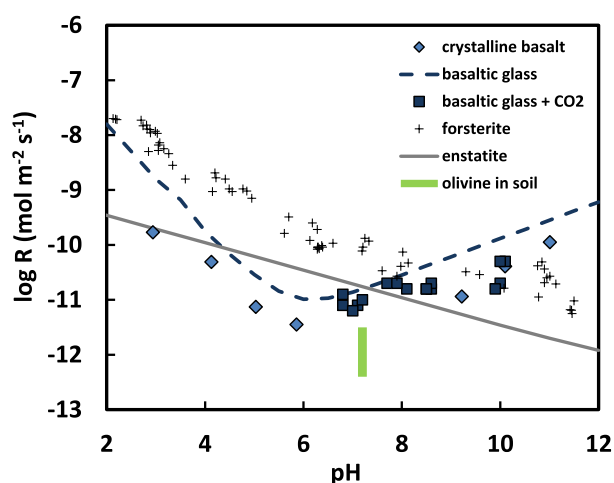


Fig. 1. BET normalized dissolution rate of basalt, forsterite and enstatite (based on Silica release) as a function of pH at 25 °C. Crystalline basalt (Gudbrandsson et al., 2011), basaltic glass calculated from (Gislason and Oelkers, 2003), basaltic glass + CO<sub>2</sub> (Stockmann et al., 2011), forsterite (Pokrovsky and Schott, 2000), enstatite (Oelkers and Schott, 2001) and olivine, added to soil (Renforth et al., 2015).

Mg<sup>2+</sup> and Fe<sup>2+</sup> dominantly released from pyroxenes and olivine a linear decrease was observed (Gudbrandsson et al., 2011). The reactivity of basalt was not hindered by the formation of secondary carbonates in the experiments carried out by Stockmann (2011). Basaltic glass and its crystalline counterparts exhibit a similar pH dependence on dissolution rates, which decrease with increasing Silicon content of the solid phase. Wolff-Boenisch et al. (2006) investigated the influence on crystallinity on dissolution rates of silicate minerals and their amorphous counterparts at pH 4 and found that crystallinity influences the dissolution rate for Silicon rich phases, whereas ultramafic glasses dissolve similar to their crystalline counterparts. In general, dissolution rates of volcanic rocks decrease with increasing Silicon content (Wolff-Boenisch et al., 2004).

To understand better the importance of mafic and ultramafic rocks as reactants in the long term global Silicon Carbon cycle, basalt weathering rates have been estimated at catchment scale in several studies (e.g. Babechuk et al., 2014; Das et al., 2005; Dessert et al., 2001; Li et al., 2016; Louvat and Allègre, 1997; Moulton et al., 2000). However, reconciling the observations from field scale with the laboratory derived weathering rates remains a challenge. In general the difference in laboratory and field rates can be attributed to both, physical i.e. hydrological features (Velbel, 1993), chemical features such as surface passivation though leached layer formation or the precipitation of secondary phases (Daval et al., 2018; Velbel, 2009), or a mixture of both, such as a long fluid residence times. The prediction of surface passivation itself is not straightforward. For instance the formation of passivating layers may be prevented through fungal biofilm, thereby increasing dissolution rates (Gerrits et al., 2020). On the other hand, the passivating effect of Al on the solubility and dissolution rate of amorphous silica and quartz has been investigated by (Bickmore et al., 2006; Iler, 1973), showing that Al containing layers not only reduce dissolution rates but also apparent overall solubility. Fluid residence time, soil moisture and mineral/fluid ratios are crucial when investigating the rate of chemical weathering (Maher, 2010; Navarre-Sitchler et al., 2011; White and Brantley, 2003). The complex interrelationship between gas and water in the unsaturated zone further complicates the calculation of realistic weathering rates (Harrison et al., 2015, 2017). In addition, field weathering rates are influenced by trace metals (Oelkers et al., 2018), bacterial communities (Wild et al., 2019) or organic ligands (Peréz-Fodich and Derry, 2019; Perez et al., 2015). They are not only

dependent on a specific surface area but can be influenced by crystallographic orientation of minerals (Daval et al., 2013).

In the context of enhanced weathering, dissolution of rock powder added to soil cannot be calculated merely based on dissolution rates. The shrinking core model has been used to describe the time for complete dissolution of a grain (Hangx and Spiers, 2009). The authors estimated that the addition of olivine to seawater requires particle sizes <10 µm to achieve relatively complete dissolution of 100 years. With a similar approach Renforth et al. (2015) estimated that the particle size of olivine added to soil, would have to be in the range of 0.1–0.01 µm to dissolve within 5 years.

Notwithstanding the uncertainties related to field weathering, in this study dissolution was calculated, using a dissolution rate from (Gudbrandsson et al., 2011), who measured a BET surface (Brunauer et al., 1938) normalized rate of  $3.55 \cdot 10^{-12} \text{ mol m}^{-2} \text{ s}^{-1}$  at pH 5.84 and 25 °C (see Fig. 1). This is probably at the high end of weathering rates, given the discrepancy between lab and field rates and 25 °C is certainly far away from the annual average temperature in Austria. It is however close to the high end of dissolution rates reported for olivine in soil (Renforth et al., 2015) (see Fig. 1), and it is also in the range of the elemental release rates reported for basalt added to soil (Kelland et al., 2020). These authors measured an elemental Mg release rate of  $6.6 \cdot 10^{-13} \text{ mol m}^{-2} \text{ s}^{-1}$  and a Ca release of  $2.8 \cdot 10^{-12} \text{ mol m}^{-2} \text{ s}^{-1}$ . For comparison, the stoichiometric dissolution of our model basalt (Table 1) at the chosen rate gives an elemental release rate for Mg of  $3.54 \cdot 10^{-13} \text{ mol m}^{-2} \text{ s}^{-1}$  and  $1.06 \cdot 10^{-12} \text{ mol m}^{-2} \text{ s}^{-1}$  for Ca.

The BET surface area of the model basalt was calculated using an empirical equation, provided in (Brantley and Mellott, 2000) for olivine, where  $d$  is the particle diameter.

$$\log \text{SSA} (\text{cm}^2 \text{g}^{-1}) = 5.3 - 1.1 \cdot \log(d) \quad (4)$$

Molecular weight of the basalt is  $125 \text{ gmol}^{-1}$  and the assumed density is  $3 \text{ g cm}^{-3}$ .

#### 2.4. A modified shrinking core model

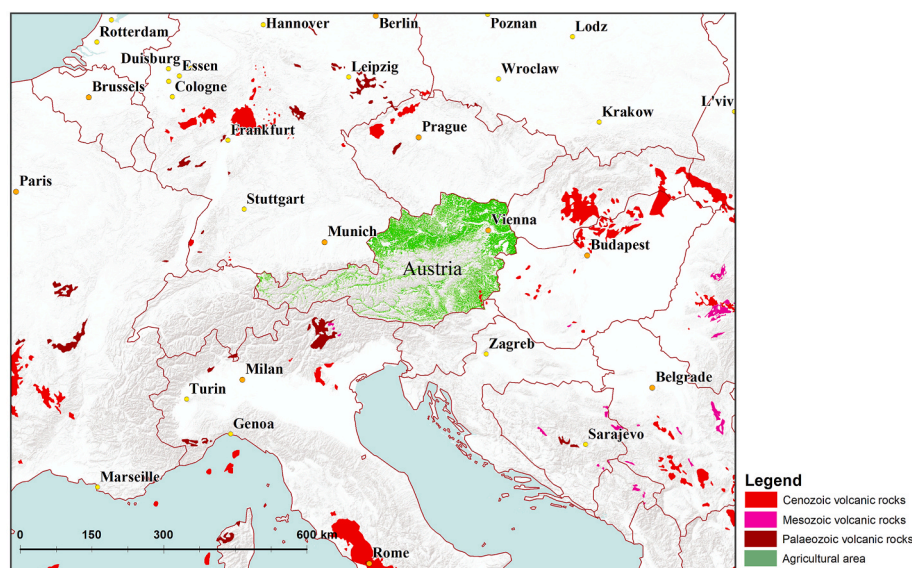
The shrinking core model (Hangx and Spiers, 2009) assumes spherical grains to model the dissolution process, which accordingly only approximates surface-volume ratios of natural grains. For this reason a steady state model, i.e. a scenario in which the specific surface area does not change over time, has been used in Strefler et al. (2018). To circumvent potential shortcomings of both approaches we include the roughness ratio ( $\text{SSA}_{\text{BET}}/\text{SSA}_{\text{Geo}}$ ) into the shrinking core model, where  $\text{SSA}_{\text{BET}}$  refers to the specific surface area calculated from gas adsorption according to Brunauer et al. (1938) and  $\text{SSA}_{\text{Geo}}$  refers to the specific surface area, based on spherical particles according to the equation provided in (Tester et al., 1994). BET normalized dissolution rates are recalculated to the geometric surface area of spherical grains of each fraction (see the supplementary file for details about the calculation). This way the overestimation of the amount of dissolution (steady state) as well as underestimation (applying BET normalized dissolution rates to a shrinking core model) can be avoided (see Fig. 4 and discussion).

#### 2.5. Geographical framework and model parameters

##### 2.5.1. Availability of volcanic rocks in Austria and neighboring countries

Potential occurrences of volcanic rocks in the vicinity of Austria comprise units from the Western and Central European volcanic province in the Massif Central (Lustrino and Wilson, 2007), the Bohemian Massif and Eger Graben (Ulrych et al., 2011), Germany (Jung et al., 2005; Jung and Masberg, 1998), the Pannonian-Carpathian Volcanic province – parts of which are based in SE Austria (Ali et al., 2013; Downes et al., 1995; Lukács et al., 2018; Seghedi et al., 2004) and the Veneto Volcanic province in Northern Italy (Peccerillo, 2005) as well as some occurrences in the Dinaric alps (Prelević et al., 2005), (see Fig. 2).





**Fig. 2.** Map showing the distribution of agricultural land in Austria and potential volcanic rocks in the wider vicinity. Layers: World terrain basemap (Sources: Esri, USGS, NOAA); World cities (Sources: Esri; Bartholemew and Times Books; U.S. Central Intelligence Agency (The World Factbook); International Organization for Standardization; United States Department of State, Bureau of Intelligence and Research; GeoNames; Executive Secretary for Foreign Names - U.S. Board on Geographic Names; U.S. National Geospatial-Intelligence Agency; BBC News; Global Mapping International); CORINE land cover 2018 (© Umweltbundesamt & European Union, Copernicus Land Monitoring Service 2018, European Environment Agency (EEA), with funding by the European Union); © EuroGeographics for the administrative boundaries; volcanic rocks from IGME-5000 (Asch, 2005). The map was created using ArcGIS® software by Esri. ArcGIS® and ArcMap™ are the intellectual property of Esri and are used herein under license. Copyright © Esri. All rights reserved.

Relatively large volcanic provinces of Permo-Carboniferous origin appear south of the Insubric line, mostly comprising intermediate to felsic volcanic rocks, e.g. dacite, trachyte, rhyolite (Cortesogno et al., 1998). The same is true for relatively large volcanic fields close in the West Carpathians, such as the central Slovakian volcanic field, mostly comprising andesite (Chernyshev et al., 2013). Similarly, volcanic rocks of the roman volcanic province are largely of intermediate composition (Beccaluva et al., 1991). (Plata et al., 2021) investigated the potential of andesitic and dacitic mining waste as a soil fertilizer, showing that soil amelioration is not limited to volcanic rocks with a low Silica content. However, Cenozoic volcanic rocks of ultramafic to mafic composition can be found in all volcanic provinces surrounding Austria (see Fig. 3). The impression gained from Fig. 3 also holds when compared to the more extensive database of volcanic rocks provided in (Lustrino and Wilson, 2007), or with respect to the data provided by (Hartmann and Moosdorf, 2012), who calculated the percentage for basic volcanic rocks of  $\approx 59.5\%$ , with intermediate compositions contributing  $8.5\%$  and acid compositions covering an area of  $32\%$  for whole Europe. However, for Central Europe, basalt may not be main rock source and other mafic volcanic rocks such as basanite or trachybasalt might need to be included to provide the enormous amounts of rock required. As noted above, differences in mineralogical and chemical composition are both crucial for the value as a fertilizer as well as dissolution kinetics. Accordingly, when we refer to basalt in this manuscript it should be considered as representative for a range of ultramafic and mafic volcanic rocks, which warrant closer examination in the future.

### 2.5.2. Agricultural areas in Austria

Cropland area covered approximately 1.325 M ha in 2019 (Statistik Austria, 2020), and additional 0.576 M ha were categorized as intensive grassland (Statistik Austria, 2018), assumed to be easily available for inorganic fertilizer application. Out of 1152 representative soil samples  $38\%$  have a soil pH  $> 7$ , in  $53\%$  of samples the pH ranges from 5 to 7 and  $9\%$  are characterized as strongly acidic (pH = 4–5). For grass land only  $6\%$  show a pH  $> 7$ . The pH of  $55\%$  of the samples falls in the range of 5–7, and  $39\%$  of the samples are between pH 4 and pH 5 (Schwarz and Freudenschuss, 2004). Soil amelioration is mostly proposed for acidic soils. However, from a perspective of CO<sub>2</sub> sequestration, soil pH has relatively low impact as long as basalt is used. The dissolution rate of basaltic glass in the relevant range (4–8.5) reaches a minimum around pH 6 (see Fig. 1). Compared to this minimum, dissolution rate is  $\approx 3$  times faster at pH 5 and also  $\approx 3$  times faster at pH 8. A pronounced pH effect would only be expected for very acid conditions – at pH 4 the

dissolution rate is  $\approx 17$  times that of pH 6 (Gislason and Oelkers, 2003). From this perspective basaltic soil amelioration could also be applied to lime rich alkaline soils, where it could provide a potassium source, decreasing the need for conventional fertilizers. In summary, an area of 1.902 M ha (cropland plus intensive grassland) are assumed to be a reasonable maximum area of application and will be used in further modeling exercises.

### 2.5.3. Mining, transport and comminution

The energy demand related to crushing and grinding of material in order to reach particle sizes small enough to achieve desirable weathering rates in soil are considered and important drawback of EW (Gerdemann et al., 2007; Moosdorf et al., 2014; Renforth et al., 2015; Rigopoulos et al., 2018b). To evaluate the interrelationship between CO<sub>2</sub> emissions from comminution and drawdown rate of CO<sub>2</sub>, three scenarios with different grain size distribution are used for modeling. The selected particle size distribution ranges from 100 to 0.1  $\mu\text{m}$  ( $< 100 \mu\text{m}$ ), 10–0.1  $\mu\text{m}$  ( $< 10 \mu\text{m}$ ) and 1–0.1  $\mu\text{m}$  ( $< 1 \mu\text{m}$ ). The particle size distribution was calculated for fixed classes, with normal distribution with a  $\mu$  of 50, 5 and 0.5 and  $\sigma$  of 30, 3 and 0.3, respectively. The resulting grain size distribution can be found in the supplementary file (Table S1). Comminution, mining and application can be considered static and resulting input parameters used for calculation of the CO<sub>2</sub> balance are listed in Table 2. Based on the relatively widespread availability of mafic and ultramafic volcanic rocks (see above) the average distance from mine to field was estimated about 300 km. This is an optimistic approach, assuming that the quarries closest to the Austrian border are also able to produce the required amount and quality. Resulting transport related CO<sub>2</sub> emissions considered in this study, are  $4.8 \text{ kg CO}_{2\text{e}} \text{ t}^{-1}$ ,  $18.9 \text{ kg CO}_{2\text{e}} \text{ t}^{-1}$  and  $59.4 \text{ kg CO}_{2\text{e}} \text{ t}^{-1}$ , for transport by rail, a  $< 7.5$  ton truck and 20–40 ton truck, respectively. Total CO<sub>2</sub> emissions including mining, comminution, transport and application calculated on base of 300 km transport are shown in Table 3 for three scenarios, using different grain size distributions ( $< 100 \mu\text{m}$ ,  $< 10 \mu\text{m}$ ,  $< 1 \mu\text{m}$ ).

## 3. Results and discussion

### 3.1. Assessment of the modified shrinking core model

Strefler et al. (2018) used a steady state model to estimate the dissolution of rock powder. Such a model assumes that dissolving powder is always present in the same amount and accordingly the

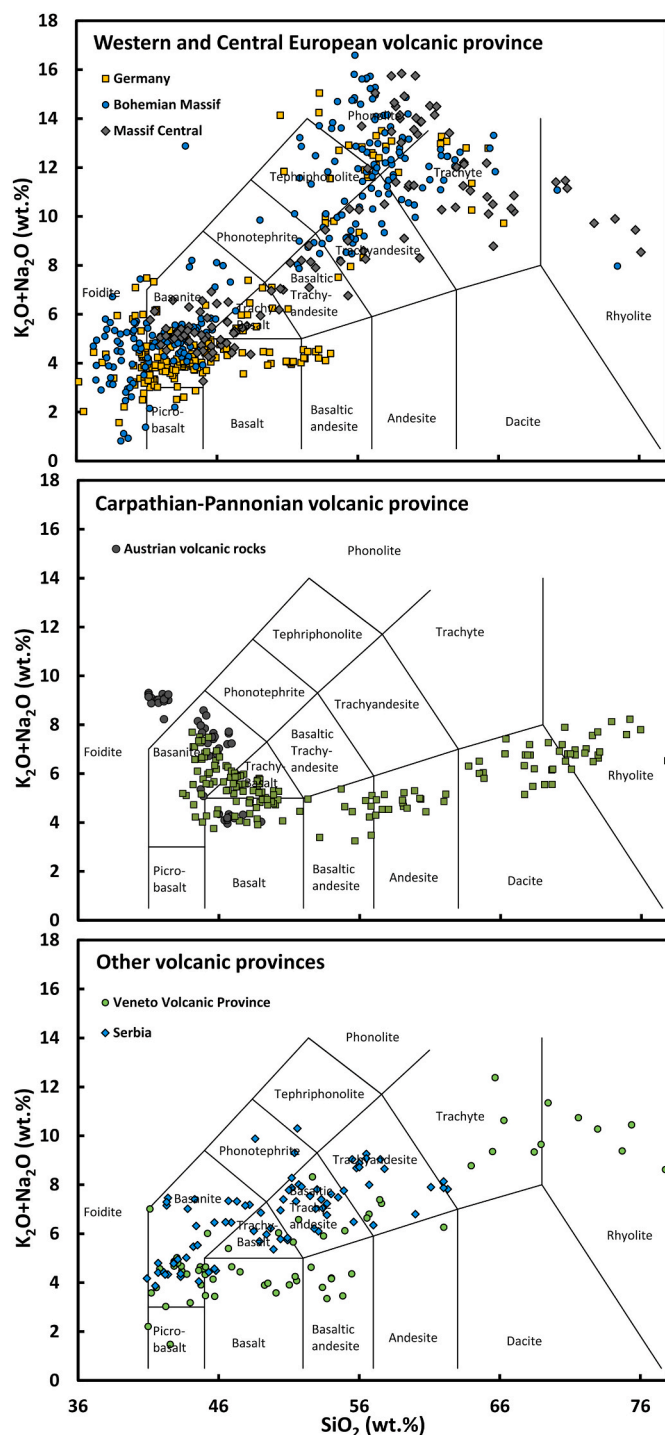


Fig. 3. Total alkali content ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ) versus  $\text{SiO}_2$  of selected volcanic rocks in the wider vicinity of Austria. (a) Carpathian – Pannonian region; data from (Ali et al., 2013; Ali and Ntafos, 2011; Harangi et al., 2007, 2015, 2007; Lukács et al., 2018), (b) Western and Central European volcanic province (Alibert et al., 1983; Briot et al., 1991; Chauvel and Jahn, 1984; Dautria et al., 2004; Haase et al., 2004, 2017, 2004; Hegner et al., 1995; Jung et al., 2006; Jung and Masberg, 1998; Mertes and Schmincke, 1985; Riley et al., 1999; Ulrych et al., 2003, 2011, 2003; Wilson et al., 1995; Wörner and Schmincke, 1984); (c) Other volcanic regions (Macera et al., 2003; Milani et al., 1999; Prelević et al., 2005).

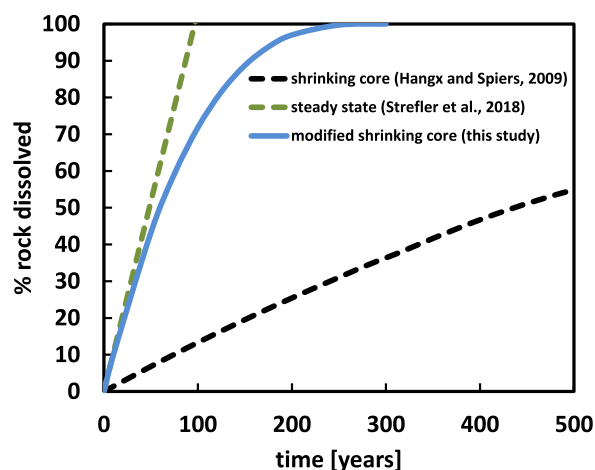


Fig. 4. Dissolution of basalt powder with a grain size of  $20\ \mu\text{m}$  and an of  $\text{SSA}_{\text{BET}}$  of  $0.74\ \text{m}^2\ \text{g}^{-1}$  at a rate of  $3.55\ 10^{-12}\ \text{mol}\ \text{m}^{-2}\ \text{s}^{-1}$  calculated after Hangx and Spiers (2009), Strefler et al. (2018) and this study.

Table 2

Input parameters, used for the calculation of the  $\text{CO}_2$  balance.

	$\text{kg}\ \text{CO}_2\ \text{kWh}^{-1}$	$\text{kg}\ \text{CO}_2\ \text{e}\ \text{t}^{-1}\ \text{km}\ \text{kWh}\ \text{t}^{-1}$	$\text{kg}\ \text{CO}_2\ \text{t}^{-1}$
electricity generation (Austria) <sup>a</sup>	0.26		
Road transport ( $<7.5\ \text{t}$ ) <sup>b</sup>		0.198	
Road transport ( $20\text{--}40\ \text{t}$ ) <sup>b</sup>		0.063	
Railway transport <sup>b</sup>		0.016	
comminution ( $<100\ \mu\text{m}$ ) <sup>c</sup>			18.9
comminution ( $<10\ \mu\text{m}$ ) <sup>d</sup>			173.0
comminution ( $<1\ \mu\text{m}$ ) <sup>e</sup>			556.0
Mining <sup>e</sup>			7.0
application low estimate <sup>e</sup>			1.0
application high estimate <sup>e</sup>			4.0

<sup>a</sup> <https://secure.umweltbundesamt.at/co2mon/co2mon.html>.

<sup>b</sup> (Schmied and Knörr, 2013).

<sup>c</sup> Energy requirement for comminution to  $<100\ \mu\text{m}$  from the table of bond work index (<https://www.911metallurgist.com/blog/table-of-bond-work-index-by-minerals>).

<sup>d</sup> (Hangx and Spiers, 2009).

<sup>e</sup> (Moosdorf et al., 2014).

Table 3

Total  $\text{CO}_2$  emissions in  $\text{kg}\ \text{t}^{-1}$  of rock, including mining, transport (300 km), comminution and application.

Grain size	Railway transport	Road transport ( $20\text{--}40\ \text{t}$ )	Road transport ( $<7.5\ \text{t}$ )
$<100\ \mu\text{m}$	17.7	31.8	75.3
$<10\ \mu\text{m}$	57.8	71.9	115.4
$<1\ \mu\text{m}$	157.4	171.5	215.0

surface area available for water rock interaction does not change over time. They used a  $20\ \mu\text{m}$  powder with a  $\text{SSA}_{\text{BET}}$  of  $1.69\ \text{m}^2\ \text{g}^{-1}$  and yielded a dissolution of 19.6% within 1 year. In general our scenario is relatively conservative compared to that. With a  $\text{SSA}_{\text{BET}}$  of  $0.74\ \text{m}^2\ \text{g}^{-1}$  and a lower dissolution rate of  $3.55\ 10^{-12}\ \text{mol}\ \text{m}^{-2}\ \text{s}^{-1}$  the steady state model would result in only 1.03% of the powder dissolved within 1 year. Interestingly, using our modified shrinking core model, we calculate a similar amount of 1.03%. Apparently, accounting for the shrinking core is not very important during initial stages of dissolution and a steady state approach seems valid even with the 19.6% dissolved in Strefler

et al. (2018) (see Fig. 4). However, as the dissolution of the basalt powder continues over several years, a steady state model becomes increasingly unrealistic. The complete dissolution according steady state assumption takes 97 years, in which case only 70% of the powder are dissolved using our modified shrinking core (see Fig. 4). The shrinking core as originally proposed in Hangx and Spiers (2009) dissolves only 0.14% of powder within 1 year and the complete dissolution takes 2120 years. This underestimation is however a result of applying a BET normalized dissolution rate to the geometric surface of a sphere and simple recalculation can circumvent this problem (see supplementary file for details). Accordingly, we propose the shrinking core model applied in this study as a plausible way to estimate the dissolution of rock powder.

### 3.2. Influence of particle size on weathering rates

Based on Eq. (4) the average  $SSA_{BET}$  resulting is 10.53 ( $<100\ \mu\text{m}$ ), 18.61 ( $<10\ \mu\text{m}$ ) and  $67.42\ \text{m}^2\ \text{g}^{-1}$  ( $<1\ \mu\text{m}$ ), respectively. Time for complete dissolution of the powder was calculated to be 1696, 134 and 10.7 years for the  $<100\ \mu\text{m}$ ,  $<10\ \mu\text{m}$  and  $<1\ \mu\text{m}$  powder, respectively. However, as the reservoir gets depleted and the grains become increasingly small, dissolution becomes relatively ineffective and for instance 87% of a  $100\ \mu\text{m}$  grain will have dissolved in half the time, needed for complete dissolution. Accordingly, the evaluated timeframe in Table S1 and Fig. 5 is for 97.5% dissolution in which case the dissolution takes 1205, 96 and 7.6 years. As this time is based on the largest size grains in each powder, smaller grains will dissolve completely and based on the chosen grain size distribution  $>99.9\%$  of the basalt powder will dissolve within the chosen timeframe (see Fig. S1 in the supplementary file).

The potential  $\text{CO}_2$  drawdown associated with the dissolution of the respective powder is shown in Fig. 5a–c for the first 10 years and  $R_{\text{CO}_2\ \text{max}}$  and  $R_{\text{CO}_2\ \text{low}}$ . Based on their experiments, Kelland et al. (2020) modeled a cumulative removal of  $\text{CO}_2$  of  $44\ \text{kg}\ \text{CO}_2\ \text{t}^{-1}$  of basalt within 5 years through the application of basalt with a diameter of  $<128\ \mu\text{m}$  in good agreement with our calculation for the  $<100\ \mu\text{m}$  (Fig. 5a) which yields a drawdown  $35\ \text{kg}\ \text{CO}_2\ \text{t}^{-1}$  after 5 years. Within the chosen observation period, increasing the reaction rate through decreasing the grain size is decisive in the  $R_{\text{CO}_2\ \text{low}}$  and the  $R_{\text{CO}_2\ \text{max}}$  scenario (see Fig. 6). Apparently comminution, despite its high energy demand is not the controlling factor on the  $\text{CO}_2$  drawdown. On the contrary, given the slow dissolution of the  $<100\ \mu\text{m}$  fraction (1698 years for complete dissolution), even best practice (rail transport) will only contribute limited to climate change mitigation (see Table S2 and Fig. 6), and in the case of poor logistics (truck transport  $<7.5\ \text{t}$ ) it could take  $\approx 40$  years until the  $\text{CO}_2$  balance reaches at least net 0. However, under a  $R_{\text{CO}_2\ \text{max}}$  scenario, considerable  $\text{CO}_2$  drawdown could be reached even with a relatively large particle size. It can be argued that  $R_{\text{CO}_2\ \text{max}}$  is high and has not been proposed by any of the other studies in context with basalt application. The exact mechanisms of  $\text{CO}_2$  drawdown during basalt weathering and the corresponding  $R_{\text{CO}_2\ \text{max}}$  are still unclear (see above) but in any case the assumed drawdown is close to the drawdown for e.g. olivine, showing the potential within ultramafic rocks in general. We note that grinding down to  $<1\ \mu\text{m}$  is to our knowledge technically not possible at this moment at industrial scale. Notwithstanding the practical implications of the high energy demand (see section 3.3) it goes to show however that the smallest technically possible grain size should be aimed at.

### 3.3. Cumulative $\text{CO}_2$ removal in Austria

Assuming the maximum application on both intensive grassland ( $0.576\ \text{M ha}$ ) and agricultural land ( $1.326\ \text{M ha}$ ), we estimate the amount of  $\text{CO}_2$  drawdown potential for the application of  $10\ \text{kg rock m}^{-2}$  ( $100\ \text{t ha}^{-1}$ ). The cumulative amount of basalt applied in this case is  $190\ \text{Mt}$ . Using rail transport and  $R_{\text{CO}_2\ \text{low}}$ , this amounts to a removal of

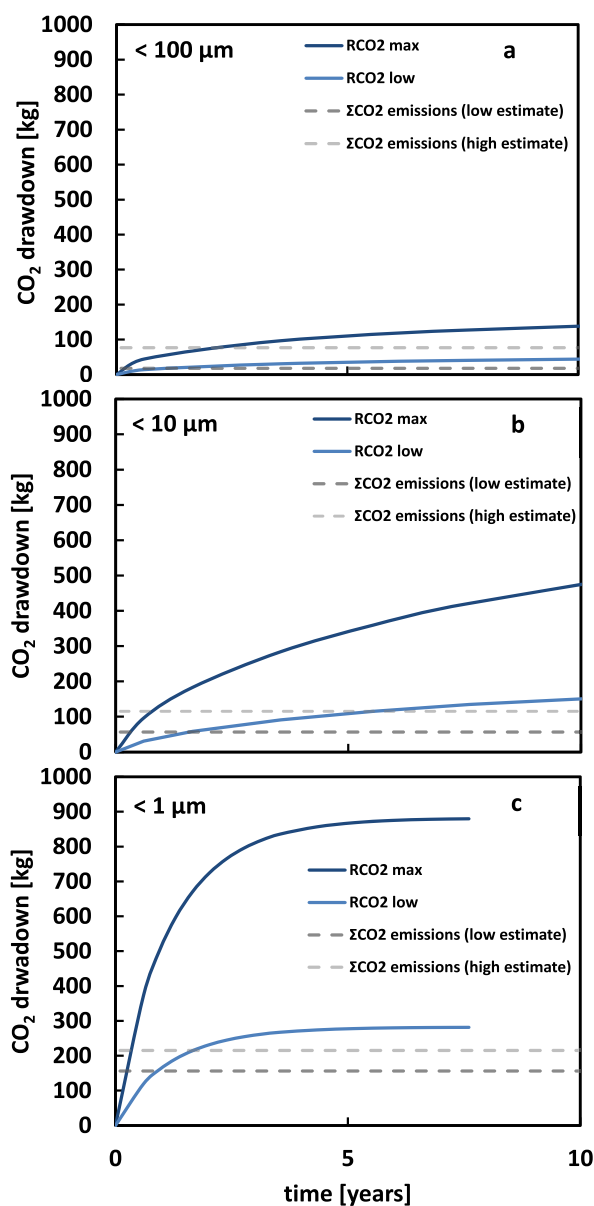


Fig. 5. Evolution of  $\text{CO}_2$  drawdown in  $\text{kg t}^{-1}$  of basalt, using  $R_{\text{CO}_2\ \text{low}}$  of  $282\ \text{kg}$  and  $R_{\text{CO}_2\ \text{max}}$  of  $880\ \text{kg}$ . Grey dashed lines denote the estimates for  $\text{CO}_2$  emissions, generated through mining, grinding, transport (300 km) and application; the low estimate is based on railway transport; the high estimate is based on road transport using a  $<7.5\ \text{t}$  truck.

5.2 and  $18.3\ \text{Mt}\ \text{CO}_2$  for the  $<100$  and  $<10\ \mu\text{m}$  fraction, respectively. The annual output of greenhouse gases in Austria amounts to roughly  $80\ \text{Mt}\ \text{CO}_2\text{e}$  (Klimaschutzbericht, 2019). This translates to a net removal of 6.5% and 22.9% of Austria's annual greenhouse gas emissions ( $\text{CO}_2\text{e}$ ) within a 10 year cycle (see Table S2). It is important to note however, that current transportation practices could easily turn this into a positive emission technology (see Fig. 6a and Table S2). For  $R_{\text{CO}_2\ \text{max}}$  the potential drawdown would be tremendous from 29.2% ( $<100\ \mu\text{m}$ ) up to a maximum of 100.5% ( $<10\ \mu\text{m}$ ) of Austria's annual greenhouse gas emissions ( $\text{CO}_2\text{e}$ ) within a 10 year cycle. This is in good agreement with the study of (Moosdorf et al., 2014), who used a comparable  $R_{\text{CO}_2}$  to evaluate the potential of ultramafic rocks and found that on a global scale transport would not influence  $\text{CO}_2$  drawdown in a way which makes the application unfeasible. We note however that including reaction rates, the gross  $\text{CO}_2$  removal after 10 years in our estimate is  $140$



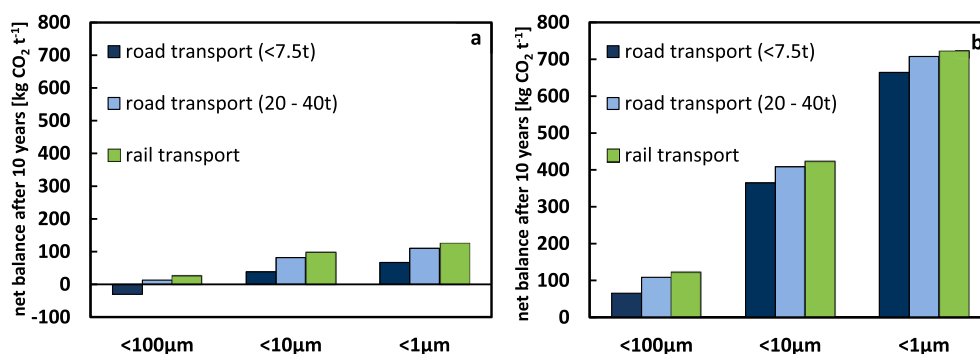


Fig. 6. CO<sub>2</sub> balance after 10 years for different scenarios (see Table S2), and R<sub>CO<sub>2</sub> low</sub> of 282 kg CO<sub>2</sub> t<sup>-1</sup> rock (a) and R<sub>CO<sub>2</sub> max</sub> of 880 kg CO<sub>2</sub> t<sup>-1</sup> rock (b).

(<100 µm) and 480 (<10 µm) kg CO<sub>2</sub> t<sup>-1</sup>. The range of 500–1000 kg CO<sub>2</sub> t<sup>-1</sup> obtained for complete dissolution is misleading because it hides the timeframe needed to achieve such drawdown.

### 3.4. A global perspective on the cumulative CO<sub>2</sub> removal

In a previous study Streffer et al. (2018) considered a global area of 7.9 M km<sup>2</sup> for application of rock powder (15 kg m<sup>-2</sup>), and estimated a drawdown of 4.9 Gt CO<sub>2</sub> a<sup>-1</sup>. We note that the dissolution rate underlying this estimate is an order of magnitude higher than ours. Using the modified shrinking core model, 6.3% of the <100 µm powder dissolve within one year, compared to 19.6% in Streffer et al. (2018). The catchment conversion equation provided by those authors yields a drawdown of 1.65 Gt CO<sub>2</sub> a<sup>-1</sup>. This number increases to 4.0 Gt CO<sub>2</sub> a<sup>-1</sup> for powders with grain size <10 µm, and to 15.4 Gt CO<sub>2</sub> a<sup>-1</sup> for powders with grain size <1 µm, respectively. These numbers however refer to the gross CO<sub>2</sub> drawdown. Including the emissions from the 300 km railway scenario, mining, comminution and application the drawdown roughly balances the emissions during the first year (see Fig. 7). Accordingly, the actual net drawdown is much lower. After 10 years 16% of the powder (<100 µm) is dissolved. As a result the cumulative net CO<sub>2</sub> drawdown after 10 years will only be 2.54 Gt, i.e. 0.254 Gt CO<sub>2</sub> a<sup>-1</sup>. If applied annually over the course of 10 years, the CO<sub>2</sub> drawdown increases from approximately zero in the first year to a cumulative removal of 3.2 Gt CO<sub>2</sub> a<sup>-1</sup> in year 10. The annual application on a global area of 7.9 M km<sup>2</sup> at the suggested rate of 15 kg m<sup>-2</sup> sums up to 118.5 Gt, and this amount would have to be added every year in order to reach the cumulative 3.2 Gt CO<sub>2</sub> a<sup>-1</sup>. The large volume of rock needed in this scenario is in conflict with the relatively low annual application rate of 3 Gt a<sup>-1</sup> of basalt to sequester 1 Gt CO<sub>2</sub> a<sup>-1</sup>, proposed in Streffer et al. (2018). But, other than suggested through a steady state model, it is not possible to simply replenish the amount of dissolved powder, considering how weathering differs depending on grain size. Smaller grains dissolve first, as the larger ones stay, which do not offer as much exposed surface.

A recent estimate applied 5.9 Gt of rock per year to reach the goal of 1 Gt CO<sub>2</sub> a<sup>-1</sup> using a powder with grain size of <100 µm (Beerling et al., 2020). The reactive transport model in this study contains a range of parameters such as soil pH or biological impacts that are not considered in our assessment. Nevertheless it is possible to compare the two approaches. At our given R<sub>CO<sub>2</sub></sub> of 0.282 t CO<sub>2</sub> t<sup>-1</sup> of rock, reaching a drawdown of 1 Gt CO<sub>2</sub>/5.9 Gt of basalt requires that 59% of the basalt dissolve. Within the timeframe of 10 years the respective dissolution is 16% (<100 µm). To achieve the required drawdown with the modified shrinking core model we would have to apply a dissolution rate of basalt of 5.5 10<sup>-11</sup> mol m<sup>-2</sup> s<sup>-1</sup>, which is close to the dissolution rate of basalt under acidic conditions at pH 4 and 25 °C (see Fig. 1). With the respective chosen dissolution rate and including emissions from the 300 km railway scenario the annual reapplication of powder reaches a

removal of 0.16 Gt CO<sub>2</sub> a<sup>-1</sup> (<100 µm) after 10 years (see Fig. S2). Averaged over the timeframe of 10 years the drawdown is only 0.08 Gt CO<sub>2</sub> a<sup>-1</sup> (<100 µm). Using a powder (<10 µm) the dissolution after 10 years is 54% and the annual reapplication amounts to 0.57 Gt CO<sub>2</sub> a<sup>-1</sup> with an average of 0.24 Gt CO<sub>2</sub> a<sup>-1</sup>. The extent to which this accumulation effect is valid is governed by a range of criteria, including surface passivation of grains.

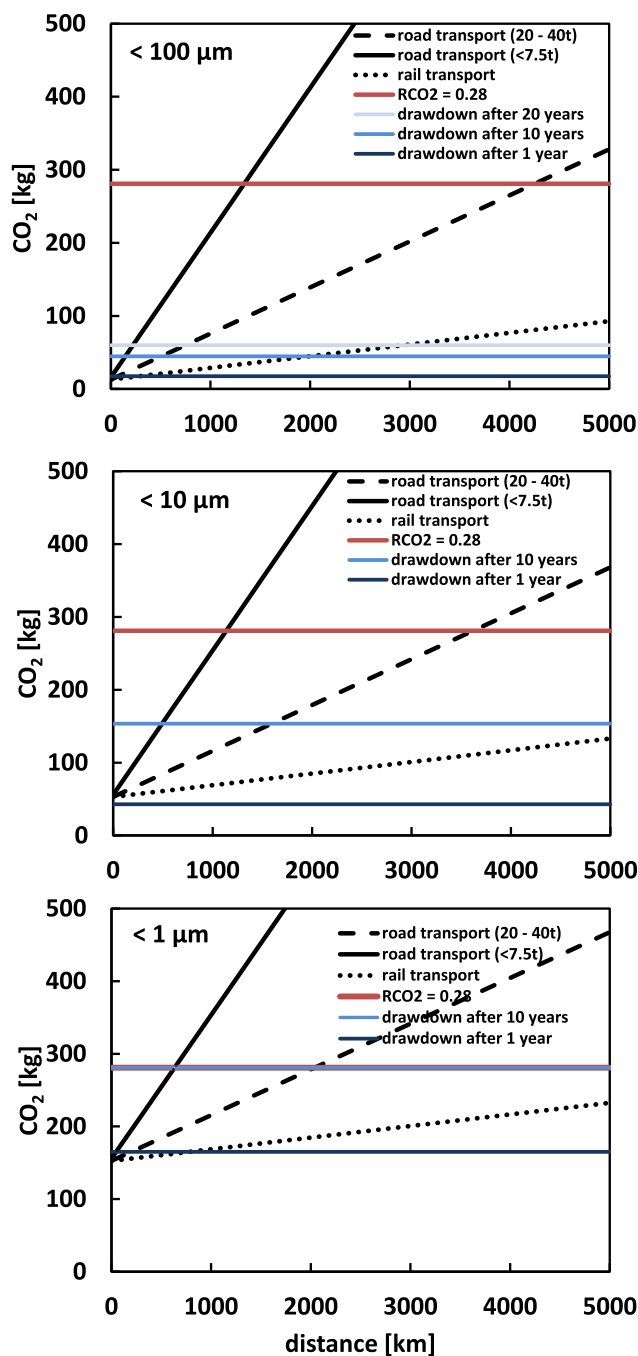
The relationship between grain size dependent weathering and transport related emissions is shown in Fig. 7. For truck transport the offsetting distances, at which potential carbon capture (R<sub>CO<sub>2</sub>min</sub> of 0,282 t CO<sub>2</sub> t<sup>-1</sup> of rock) is neutralized by transport related emissions, increase from approximately 630 km (<1 µm) to 1340 km (<100 µm). This is in good agreement with the estimates from Lefebvre et al. (2019) who calculated an offsetting distance around 990 km for enhanced weathering. But relating transport emissions to potential carbon capture does not account for the time needed to dissolve the basalt powder. Considering the drawdown after 10 years this distance decreases to 160 km (<100 µm), but it stays 630 km using the fine powder (<1 µm), which will dissolve almost completely within 10 years. Using railway transport, offsetting distances, related to potential carbon capture, decrease from 16,800 km (<100 µm) to 8000 km (<1 µm). Considering the drawdown after 10 years the offsetting distance of the <100 µm powder is still 2000 km and it stays at 8000 km for the fine powder (<1 µm).

These calculations show that it may be possible to accept larger transport distances for very fine particles, despite the high energy demand for grinding. In contrast, the use of quarry fines with a size <5 mm was evaluated by Lefebvre et al. (2019). The dissolution of a 5 mm grain according to our shrinking core model takes around 120,000 years. Not knowing the actual grain size distribution of the quarry fines, we can still hypothesize that they will be very unlikely to contribute to CO<sub>2</sub> drawdown within a reasonable timeframe, independent of the actual transport distance.

### 3.5. Energy requirement

The electricity generation in Austria amounted up to 73,460 GWh in the year 2019, and 20,901 GWh of that was generated through biogenic or fossil fuels (<https://www.e-control.at/betriebsstatistik2019>). For grinding down of 190 Mt of basalt to <10 µm and <100 µm, 32,904 GWh and 3595 GWh are needed, respectively. This is approximately 45%–4.9% of the annual power generation. Given the relatively long time for complete dissolution of the powder, it can be argued, that application would not be carried out every year and the optimal application rate is yet to be determined. Still, even if the application cycle is 10 years the energy requirement is 4.5% (<10 µm) or 0.5% (<100 µm) of annual electricity generation. A detailed analysis of the transformation and growth potential of the energy sector in Austria over the next decades is outside the scope of this study. However we note, that back until the





**Fig. 7.** Relation between grain size dependent weathering and maximum transport distances to offset potential capture. The offsetting distance decreases from 1340 km (<100  $\mu\text{m}$ ) to 634 km (<1  $\mu\text{m}$ ) for truck transport. However considering the drawdown after 10 years this distance decreases to 161 km for in the <100  $\mu\text{m}$  scenario. For rail transport the offsetting distance decreases from 16,756 km (<100  $\mu\text{m}$ ) to 8027 km (<1  $\mu\text{m}$ ). Again considering the drawdown after 10 years distance it is only 1993 km (<100  $\mu\text{m}$ ).

year 2001, the amount of electricity import was always greater than export. Reducing energy demand is considered one of the key factors in fighting climate change over the next decades and in this context liberating 0.5% of the annual electricity generation for rock comminution will not be an easy task. The use of quarry fines, waste and byproducts from the steel and cement industry as soil fertilizers could partially alleviate this problem as those materials have already been

produced in high volumes (Das et al., 2019; Manning et al., 2013; Renforth, 2019; Washbourne et al., 2012). While these potential sources warrant further investigation, large parts of historical wastes may be unavailable for re-extraction (Pullin et al., 2019) and focus should be put on management of future production. In addition, steel slag is already used as an additive to cement, thereby contributing to reducing emissions in the cement industry (Yi et al., 2012). Accordingly, tradeoffs between different forms of reuse have to be considered. We also hypothesize that the application of basalt to agricultural land as natural fertilizer holds a unique advantage compared to other NETs with respect to public acceptance. This advantage might vanish in the case of steel slag and cement waste. Finally, uncertainty in our assessment is related to the energy amount for grinding. The comminution of material to a grainsize in the range of 10  $\mu\text{m}$  could be achieved with lower energy input than assumed in prior publications, given the application of state of the art milling equipment (de Bakker, 2014; Gao et al., 2002). Accordingly, the actual amount of energy used for grinding poses an uncertainty in this study and the need to better constrain this has already been emphasized by (Hartmann et al., 2013).

### 3.6. The amount of basalt needed

With an assumed density of  $3 \text{ g cm}^{-3}$ , the 190 Mt basalt that are applied to Austrian land fill a volume of  $0.063 \text{ km}^3$  (a cube of approximately 400 m side length). The annual amount of basalt mined in Austria was around 1.5–2 Mt in the years 2015–2018 (Österreichisches Montan-Handbuch, 2019 - 93. Jahrgang, 2019). The cumulative amount of all mined rocks and minerals amounts to  $\approx 70 \text{ Mt a}^{-1}$ . The necessary amount of basalt can most likely not be sustained through mining on Austrian territory, although large volumes of volcanic rock might be found beneath the surface (Heritsch, 1982). In addition,  $100 \text{ t ha}^{-1}$  is high and it has been shown that considerable  $\text{CO}_2$  drawdown can also be achieved with lower application rates (Dietzen et al., 2018; ten Berge et al., 2012), potentially allowing to reduce the amount of basalt. In any case, the mafic lavas of the central European volcanic province in Germany comprise several thousand  $\text{km}^3$  (Jung et al., 2005) and the eruptive volume of the Vogelsberg (central Germany) alone is  $\approx 600 \text{ km}^3$  (Bogaard et al., 2003). Kereszturi et al. (2011) calculated a volume of volcanic rocks for the Bakony-Balaton Highland Volcanic Field in the Pannonian Carpathian region of  $\approx 2.9 \text{ km}^3$  and large volumes of volcanic rocks are present in the Bohemian Massif, for instance in the Doupov Mountains ( $123 \text{ km}^3$ ) (Šhrbený, 1995). Conclusively, there is potential for volcanic rock mining in central Europe for several application cycles on agricultural land, not just in Austria. However, mining even if related to sustainable agriculture and climate change mitigation inevitable is in confrontation with other aspects of environmental protection, related to the conservation of nature for ecological, economic and cultural reasons. The enormous touristic potential of some of the regions, by means of cultural heritage, the thermal spa tourism often related to Cenozoic volcanism, agricultural use in general and the high quality wines often related to volcanic terrains in particular put limits on the mining capacity of the regions. In addition, environmental impact assessment of related mining projects can be enduring. Even if a first assessment shows the enormous volumes of volcanic rocks in Central Europe, not every occurrence will provide the desired chemical composition (see Fig. 2), and high quality outcrops are already heavily exploited. Therefore if basalt fertilizer is to play a more prominent role in Austria's (and Europe's) land management practices, strategic planning of the resources use is now warranted on the European level.

## 4. Conclusion

This study assessed the potential of EW through application of basalt on agricultural land. Two alternative scenarios for basalt with high ( $\text{RCO}_{2\text{max}} = 880 \text{ kg CO}_2 \text{ t}^{-1}$ ) and low ( $\text{RCO}_{2\text{low}} = 282 \text{ kg CO}_2 \text{ t}^{-1}$ )  $\text{CO}_2$  drawdown potential were evaluated for a single application, a reaction

time of 10 years and three different grain sizes ( $<100\ \mu\text{m}$ ,  $<10\ \mu\text{m}$  and  $<1\ \mu\text{m}$ ). Using a modified shrinking core model, the  $\text{CO}_2$  drawdown in our scenarios after 10 years is lower than in previous estimates. Using a grain size ( $<100\ \mu\text{m}$ ) and  $R_{\text{CO}_2\text{low}}$ , the drawdown is only  $45\ \text{kg CO}_2$  per ton of rock. For a transport distance of 300 km the net drawdown of  $\text{CO}_2$  ranges with decreasing grain size from  $27.3\ \text{kg CO}_2\ \text{t}^{-1}$  ( $<100\ \mu\text{m}$ ) to  $125\ \text{kg CO}_2\ \text{t}^{-1}$  ( $<1\ \mu\text{m}$ ) in the  $R_{\text{CO}_2\text{low}}$  scenario, if transport is carried out by railway. The results also indicate, that road transport would turn the application of a  $<100\ \mu\text{m}$  grain size ineffective or worse, even contribute  $\text{CO}_2$  emission. However, transport becomes less important with decreasing grain size. Application of  $100\ \text{t ha}^{-1}$  with a  $<10\ \mu\text{m}$  grain size could draw down approximately 2% of Austria's annual greenhouse gas emissions. Grinding of this amount of rock, based on an application cycle of 10 years, would require up to  $\approx 5\%$  of Austria's total annual power generation. Larger grain sizes have been proposed to circumvent this problem, but our results suggest this energy will have to be available if enhanced basalt weathering is to make an impact on the  $\text{CO}_2$  budget. However, at the moment, uncertainties related to effective mechanisms of  $\text{CO}_2$  consumption during weathering hinder precise quantification. Stoichiometric dissolution of basalt including  $\text{Al}^{3+}$ , could withdraw higher amounts of  $\text{CO}_2$  ( $R_{\text{CO}_2\text{max}} = 880\ \text{kg CO}_2\ \text{t}^{-1}$ ), in which case the application of larger grain sizes ( $<100\ \mu\text{m}$ ) could be sufficient. Uncertainties regarding the actual field weathering rates of basalt powder in soil further complicate our ability to quantify the  $\text{CO}_2$  drawdown related to EW. The precise determination of these rates remains a crucial requirement towards the large scale application of EW. Further studies from laboratory to field scale will be necessary to overcome these deficiencies in order to put the available resources to optimal use within a European strategy. In addition the influence of agricultural practice on greenhouse gas emissions is increasingly put into focus in the context of climate policy, e.g. the EU LULUCF regulation (Romppanen, 2020), and soil amelioration through the application of basalt could become an integral part of soil carbon management. At last, the energy demand related to EW produces an obvious conundrum. EW is in conflict with the need to reduce energy consumption and the necessary phasing out of fossil fuel based energy. As such, EW relies on our transformation into a sustainable society with a low carbon economy and abundant low carbon power supply to contribute to climate change mitigation. It does vice versa not contribute to this transformation.

#### CRedit authorship contribution statement

**Thomas Rinder:** Conceptualization, Methodology, Software, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Christoph von Hagke:** Conceptualization, Writing – review & editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgements

We thank Sylke Hilberg for valuable comments on this manuscript. Diego Bedoya-González and Timo Kessler are thanked for their help with creation of the map. Kayla Iacovino is thanked for providing an excel sheet for plotting TAS diagrams on her website ([www.kaylaiacovino.com](http://www.kaylaiacovino.com)). We also thank Zhifu Mi and Jing-Li Fan for editorial handling of this manuscripts. 4 anonymous reviewers are thanked for their help in the improvement of this manuscript. This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2021.128178>.

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